GENERATION OF RADIOXENON ISOTOPES

Justin I. McIntyre, Sharon Pratt, Theodore W. Bowyer, Matthew W. Cooper, James C. Hayes, Tom R. Heimbigner, Charles W. Hubbard, Harry Miley, and Michael Ripplinger

Pacific Northwest National Laboratory

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ABSTRACT

With the continued population of the International Monitoring System (IMS) network and the certification of radioxenon laboratory systems, it has become necessary to calibrate and test these systems using as many of the radioxenon isotopes as possible. Several very promising techniques have been explored recently that allow for convenient production of Xe-133, Xe-131m, the short-lived isotope Xe-135, and even the difficult-to-obtain Xe-133m. IMS national and international laboratories have traditionally obtained Xe-133 from medical isotope suppliers. The activity of these medical dose samples is extremely high (~740 MBq) and requires very careful fume hood work to dilute down to appropriate levels (~10 Bq). By waiting for 10 or more half-lives it is possible to reduce the handling regime considerably, while also obtaining a strong Xe-131m signature due to its longer half-life (11.9 days). The short-lived isotope Xe-135 (9.14 hr) and the longer-lived parent nuclide Xe-133m (2.2-day half-life) are never present in these samples. To obtain these isotopes, another source or technique is required. This paper will discuss the two techniques developed and implemented at Pacific Northwest National Lab (PNNL) and show the results obtained from each technique. The first is an in-house method that allows for on-demand production of two of these isotopes, Xe-133 and Xe-135, using highly enriched uranium (HEU) and a modest flux neutron source. The second method uses a neutron activation of stable xenon.

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OBJECTIVES

The Automated Radioxenon Sampler/Analyzer (ARSA) and the Swedish Automatic Noble Gas Analyzer (SAUNA) systems were specifically designed to detect plumes of radioxenon that are a byproduct of fissioning transuranic elements (uranium and plutonium being the most relevant). The systems detect these isotopes via different collection platforms and a subsequent nuclear-detection platform that is virtually identical in form and function. This report confines itself to two in-house methods used to generate the radioxenon isotopes necessary to calibrate the nuclear detection system. The ARSA system is described in Bowyer et al., 1996; Bowyer el al., 1998; and Bowyer et al., 2002. The SAUNA system is described in Ringbom et al., 2003.

The use of radioactive xenon is instrumental for the calibration of the beta-gamma detectors in the ARSA and SAUNA systems (Reeder and Bowyer, 1998). For two of the isotopes (Xe-133 and Xe-131m) there are commercially available sources that have been successfully obtained from medical isotope production facilities. Typically costs for a vial containing 740 MBq of Xe-133 is about $15.00 and we receive a monthly shipment so that we always have Xe-133 on hand. As an added benefit, these samples of Xe-133 also contain trace quantities of Xe-131m. If the Xe-133 samples are allowed to age for several months the ratio of Xe-131m/Xe-133 becomes quite large (>>10) and it is possible to obtain the necessary detector efficiencies, energy calibration and resolutions for the Xe-131m isotope from these sources as well.

For the remaining two isotopes (Xe-133m and Xe-135) there is no such convenient source available because of the short half-lifes (2.2 days and 9.1 hours respectively). In addition Xe-133m has proven to be technically difficult to obtain in significant quantities and because it decays to Xe-133 and it is difficult to maintain significant ratios of Xe-133m/Xe-133 without a great deal of effort. In the past Xe-133m and Xe-135 have been obtained from neutron fissioning of microgram quantities of U-235 in the research reactor located on the campus of Washington State University at Pullman, Washington (Reeder et al., 2001). Shipping and handling requirements were difficult, and it was not possible to obtain the fission products from the irradiated samples for two days after irradiation. From the previous a ratio of Xe133m/Xe133 = 1/10 was the best that was obtained.

To overcome both the time delay and the administrative planning and coordination, we have developed two separate techniques here at PNNL to obtain three of the four radioxenon isotopes needed for detector calibration. The better of the two techniques used U-235, and was called the radioxenon generator (RXG). It produced very strong signatures of Xe-133 and Xe-135. The second technique used neutron activation to produce weak signatures of Xe-125m and Xe-133 and very little else. This method has however been coupled with isotopic enrichment of stable xenon isotopes and used with a nuclear research reactor by Derek Haas et al. of the University of Texas at Austin, Texas (Haas et al., 2008, these Proceedings).

RESEARCH ACCOMPLISHED

The RXG uses the fissioning of U-235 while the second method used the direct neutron activation of stable xenon isotopes. Both techniques have yielded results but due to the low neutron effluence of the neutron sources used, the neutron activation method required much higher neutron sources and an additional isotopic separations step to be made a viable method. The U-235 fission method produced adequate quantities of Xe-133 and Xe-135 to be used for the detector calibration and was considered the superior approach.

Radioxenon from Fissioning of U-235

The RXG is composed of 10.1 grams of U_3O_8 (uranium oxide) powder, with the uranium being 95% U-235. A diagram of the RXG is shown in Figure 1-A. The uranium-oxide powder is double sealed in 3-ply 3COM SBMF filter paper to prevent the oxide powder from being dispersed and to allow fission product gases to diffuse out. The sealed powder is then contained in a primary aluminum can that has several holes drilled in the sides to allow gas passage. This primary aluminum container is than wrapped in filter paper and placed in the secondary aluminum can that has an air-tight seal and a valve to keep the fission gases in. A layer of glass wool is placed on top to further prevent the spread of the uranium powder. The aluminum containment bottle was chosen both for reasons of safety

(quartz bottles may break if dropped), and low activation products. The gas samples are drawn out of the RXG using an evacuated gas bottle to volumetrically pump the fission gases out of the aluminum container.

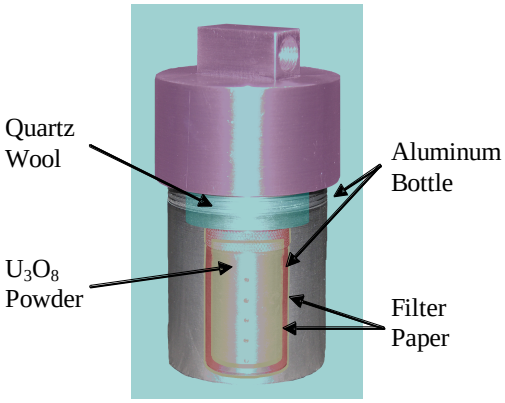

Figure 1-A. Schematic of the RXG. The overall length is 10 cm with a diameter of 4 cm.

To irradiate the uranium-oxide powder, two neutron sources were used with moderating material. During the irradiation of the sample, an 80-g PuBe source with a neutron flux of ~8.8×10^8 neutrons/s and a 10 microgram Ca-252 source with a neutron flux of 2.8×10^7 neutrons/s were placed on either sides of the RXG (see Figure 1-B). The surrounding concrete of the source storage tube aids in the thermalization of the neutrons that are generated from the two sources. To further facilitate an increase in fission reactions, two plastic bottles filled with 385 g of polyethylene beads are placed on either side of the sources as moderators to increase the flux of thermal neutrons.

The neutron irradiation time was varied over a period of hours to several weeks and produced small amounts of Xe-133m, and much larger quantities of Xe-133 and Xe-135 along with several other noble gas isotopes (Kr-85m, its daughter product Kr-85, and Kr-88). The krypton isotopes decay away quickly and do not interfere with the primary isotopes of interest. An additional follow-on effort has been undertaken at Oregon State University with Dr. David Hamby's group to use very-fast irradiation times and the high neutron effluence of their nuclear research reactor to produce favorable Xe-133m/Xe-133 ratios. In this experiment, a flow-through cell will be constructed that quickly separates out the fissioning gases from the uranium metal and other parent fission isotopes (I-133 in particular) that lead to low Xe133m/xe-133 ratios.

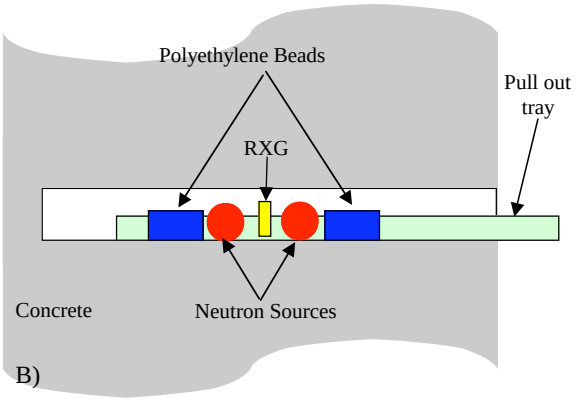

Figure 1-B. Neutron source configuration for irradiation of the RXG (not drawn to scale). The pull out tray is 5 m long. The surrounding concrete is several meters thick.

Neutron Irradiation of Stable Xenon

The irradiation of stable xenon provides another in-house technique that was pursued for the sake of availability and reduction in handling procedures. This technique used a simple plastic bottle filled with stable xenon gas to atmospheric levels. An irradiation was done using the same PuBe and Ca-252 neutron sources as was used for the uranium fissioning technique.

A quick look at the relevant isotopic abundances for stable xenon (Table 1) shows that Xe-132 and Xe-134 are the highest concentration isotopes of interest (Xe-132 + $n \rightarrow$ Xe-133 and Xe-134 + $n \rightarrow$ Xe-135) but they are not the most likely to capture a thermal neutron (*Health Physics and Radiological Health Handbook*, 1998). The neutron light isotopes of xenon (Xe-124, Xe-126 and Xe-129) are 1 to 3 orders of magnitude more likely to capture a thermal neutron. Table 1 shows the product of the thermal neutron cross-section a (column 4). It is clear from the isotopic abundance that the bulk of the strength is from the Xe-129 + $n \rightarrow$ Xe-130 reaction, which does not produce a nuclear decay. The next-highest activation is the Xe-132 + $n \rightarrow$ Xe-133 reaction followed by the Xe-124 + $n \rightarrow$ Xe-125m reaction.

Table 1. A listing of the all the stable xenon isotopes along with pertinent information for each. Clearly Xe-124 has the highest thermal neutron cross-section followed by Xe-129. The production of Xe-129m actually comes from a (*n*,*2n*) reaction on Xe-130.

Xenon Isotope	% of Atmospheric Xenon	Thermal Neutron Cross Section (mb)	Product (% * Cross Section)	Metastable Component Half-Life
Xe-124	0.10	165,000	16,500	None
Xe-126	0.09	3,500	315	None
Xe-128	1.91	480	917	None
Xe-129	26.4	22,000	580,800	8.89 days
Xe-130	4.1	450	1,845	None
Xe-131	21.4	100	2,140	11.9 days
Xe-132	26.9	500	13,450	None
Xe-134	10.4	265	2,756	290 ms
Xe-136	8.9	260	2,314	None

Table 2. Data taken from *Table of Radioactive Isotopes*, edited by E. Browne, R. B. Firestone, and V. S. Shirley, 1986.

Isotope	Half-Life	Gamma-Rays (keV)	Beta (keV)	X-Rays (keV)	CE (keV)
Xe-122	20.1 hours	148.6 (3.1%) 350.1 (7.8%)	IB 530 (<1.0%)	28–33 (78.6%)	5–24 (71%)
Xe-125	16.9 hours	188.4 (54.9%) 243.4 (28.8%)	β+ 1467 (0.69%)	28–33 (100%)	5–80 (120%) 155 (6.4%)
Xe-127	36.4 days	172.1 (23.5%) 202.9 (68%) 375.0 (15.9%)	IB 457 (<1.0%)	28–33 (54.6%)	5–33 (69.3%) 90–125 (29.4%) 138–168 (84.2%)
Xe-129m	8.89 days	39.6 (7.5%) 196.6 (4.6%)		29–35 (126.5%)	5–40 (215%) 162 (63.3%) 191–197 (60%)

Predicted activations and branching ratios indicate that four of the isotopes (Xe-122, Xe-125, Xe-127, and Xe-129m) will generate conversion electrons that may interfere with the beta spectroscopy of the radioxenon

isotopes from fission. Table 2 shows the major gamma and beta signatures that each of the four new radioxenon isotopes emit. The short-lived isotopes and metastable isotopes have not been included as they have half-lives of 3 hours or less and so are very unlikely to have much strength after generation, emission, collection, processing and eventual counting in a beta-gamma cell. Two of the isotopes are known to be produced for medical purposes (Xe-127 and Xe-122) and are routinely generated at the Los Alamos Neutron Science Center (LANSCE) in Los Alamos National Lab and the Brookhaven LINAC Isotope Producer (BLIP) at Brookhaven National Lab in New York (*Isotopes for Medicine and the Life Sciences*, 1995).

RESULTS AND ANALYSIS

Results have been obtained from both techniques as shown in this section. The fissioning of U-235 via neutrons produced the best results, and the radioactive gas has been used on both the ARSA and SAUNA systems for calibration purposes. The neutron activation method has the potential of providing very good results for individual isotopes (i.e., not a mix of three isotopes as seen from the RXG). However, it was clear from the results obtained at PNNL that a much higher neutron flux was necessary, and the use of isotopically purified xenon samples would provide unambiguous results.

Results from Fissioning of U-235

The beta-gamma spectrum obtained from the RXG is shown in Figure 2. The x-axis is the beta distribution and the y-axis is the gamma ray distributions. The upper red distribution is from Xe-135, the beta end point corresponds to 910 keV, and the gamma ray is centered at 250 keV. The next-lower distribution is from Xe-133 at a gamma energy of 80-keV and a beta end point energy of 346 keV. The lowest distribution is the 30-keV x-ray region and it contains signatures from Xe-133, Xe-133m and Xe-135. It is difficult to cleanly separate these isotopes using this region of the spectrum, and special methods of analysis are required.

A beta-gated gamma spectrum is shown in Figure 3 that clearly shows Kr-85m, Kr-88, Xe-133, and Xe-135 and is typical when the sample is immediately put into the beta-gamma detector. These isotopes were produce after a 14-day neutron irradiation of the RXG. The short half-life radiokryptons (half-lifes <5 hours) and Xe-135 have reached secular equilibrium after a couple of days of irradiation, and the long irradiation time was chosen to maximize the Xe-133 yield. There are approximately 25 mBq of Xe-133, 0.7 Bq of Xe-133m, 50 Bq of Xe-135, 10 Bq of Kr-85m, and 0.6 Bq of Kr-88 in this sample. The daughter product, Kr-85, has a much longer half-life (10.9 years) and is a beta emitter (99.6% of the time), which yields no discernable beta-gamma coincidence in the nuclear detector. From simplistic fission yield and neutron-flux-rate calculations, these values should be 3 times higher, and it is currently believed that the uranium-oxide powder is holding up most of the fission gases. To increase the yield produced by the RXG, it may be advisable to use uranium-sterate, which allows emanation of fission gases more easily.

The short-lived radiokrypton isotopes provide additional gamma and beta energy points but are not needed for the calibration of the detector. In addition, neither the ARSA nor SAUNA system ever collect or analyze for these isotopes, as the systems were designed to capture and quantify xenon-only isotopes.

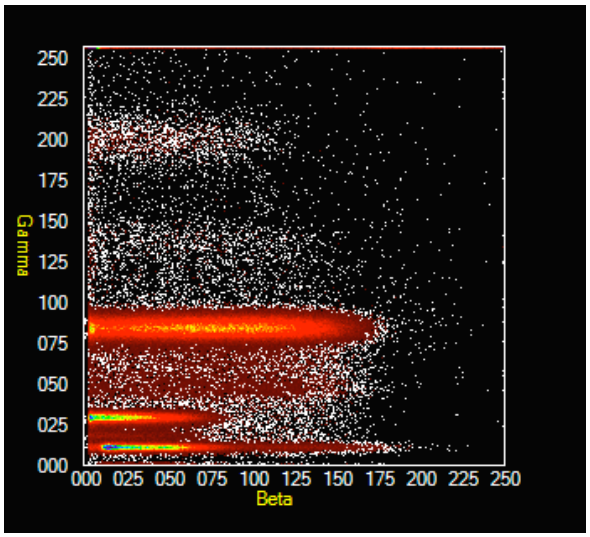

Figure 2. Beta gamma spectrum of radioxenon and radio-krypton gases produced in the RXG. There is clear indication of Xe-135 (upper distribution at gamma channel 80) and Xe-133 (middle distribution at gamma channel 30). The 30-keV x-ray region (lowest distribution at gamma channel 12) shows both Xe-133 and Xe-135 beta + conversion electron distribution. There is also just a slight amount of Xe-133m. But most of the strength for this peak (~90 %) is from Xe-133 followed by Xe-135 (~6%).

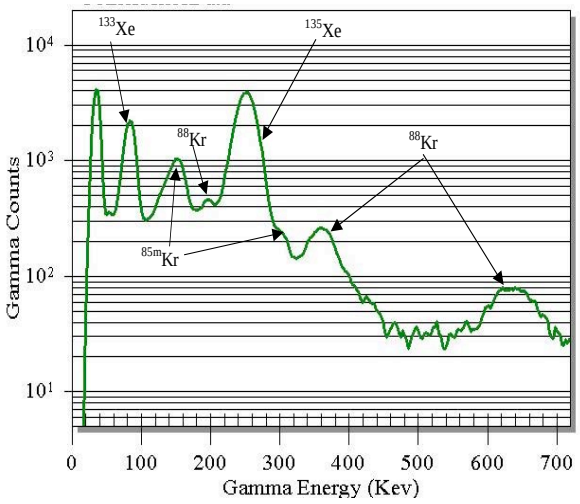

Figure 3. Beta-gated gamma spectrum of radioxenon and radio-krypton gases produced in the RXG. The peak at 30-keV (peak on the far left) contains several x-ray and gammas from several of the isotopes. Most of the strength for this peak (~90%) is from Xe-133 followed by Xe-135 (~6%).

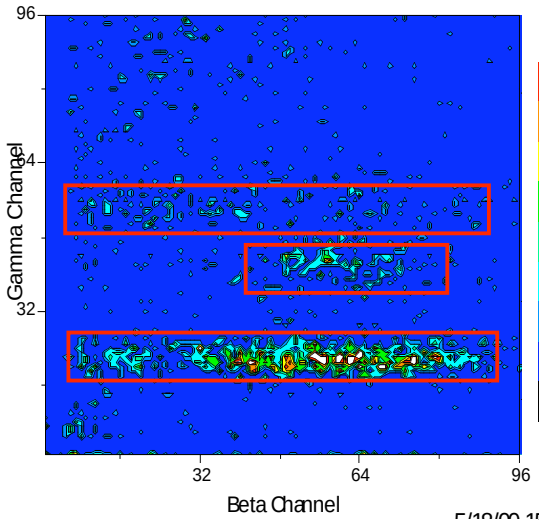

Figure 4. This plot is the beta-gamma coincidence spectrum, which shows the three distributions in question. The top box is the 80-keV gamma region, the middle is the 60-keV gamma region, and the lowest box is the 30-keV gamma region.

Results from Neutron Activation of Stable Xenon

A small flask of natural xenon (~10 cm^3) was placed next to the PuBe and Cf-252 neutron sources used in the previous U-235 irradiated RXG experiment. After 4 days of irradiation the flask was removed, and approximately 5 ml of gas was injected into a energy calibrated ARSA beta-gamma counting cells and counted for 4 days.

Figure 4 shows the beta-gamma coincidence plot, and three distributions highlighted by the red boxes can be seen. These distributions correspond to an x-ray energy of 30-keV and gamma energies of 60 and 80 keV. The 30-keV x-ray region was clearly the strongest of the three distributions and showed a definite peak along the beta axis, indicating a strong conversion electron component. The 60-keV region showed a clear peak in the beta distribution as well, with little to no characteristic beta distribution. The 80-keV distribution was clustered primarily at low energies and was indicative of the beta distribution from Xe-133.

Examination of the results after projecting the beta signal gated by appropriate γ energy cuts (Figures 5 and 6) shows the corresponding beta plus conversion electron distributions. The 1-D gamma-gated beta spectrum analysis show that the 60-keV peak is due to a Xe-125m conversion, emitting two iodine x-rays (one from the original electron capture decay and the other from the conversion process). This peak also bled into the 30-keV distribution, presumably from inefficiency of the NaI detector in detecting both the internal conversion x-ray and the conversion electron x-ray.

There seem to be two other conversion lines, corresponding to fission product radioxenon. The dataset is "sparse," however, and it is difficult to be quantitative. Comparing the 30- and 80-keV distributions, it seems likely that the 80-keV gated spectrum has a contribution from Xe-133. Most of the counts are at the lower end of the spectrum, as expected.

From these results it is clear that the activation of stable xenon will definitely complicate the analysis of the beta-gamma coincidence spectrum if large quantities are collected for a given sample. However, for calibration purposes, the technique does give several x-ray/gamma energies and several CE and beta endpoint energies.

Locally activated xenon offers a possible alternative to purchasing radioxenon from the National Institute of Standards and Technology (NIST) or producing fission product samples at the Pullman TRIGA reactor. Considering activation of Xe-124, the requisite 1-week activating and counting periods are problematic. Activating a greater amount of this isotope would require that activation would require either activation at Pullman or enriching the sample in Xe-124. Natural xenon contains only 9×10^{-4} Xe-124, and it is only observed due to its high neutron cross-section and short lifetime (17 hours). Enriching the sample to ~100% would give a signature 1,120 times more intense. Unfortunately, this isotope is expensive: 500 ml of 25% Xe-124 would cost $4,600 after shipping and handling charges. A better choice might be a sample enriched in Xe-132, as it is makes up 27% of natural xenon gas. The irradiated sample would give both β particles and a conversion line at 199 keV. This would be a useful spike sample for Xe-133 and Xe-133m ARSA and SAUNA nuclear detector tests.

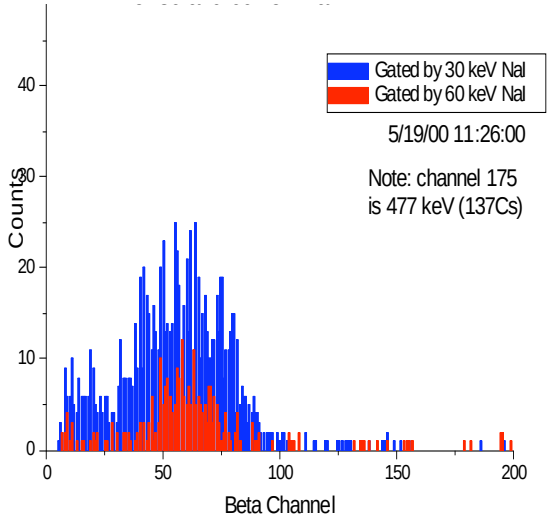

Figure 5. The projected beta spectra from the 30- and 60-keV energy cuts. Note that the 60-keV spectrum does not extend to low pulse heights, and seems to be narrower than the 30-keV plot.

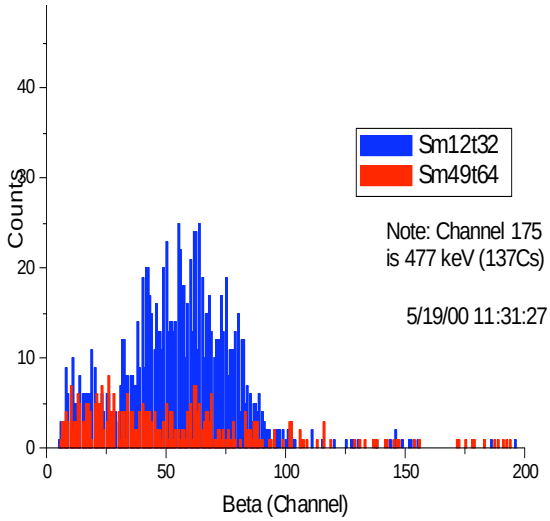

Figure 6. This plot is the comparison of beta spectra from the 30- and 80-keV NaI energy cuts. Note that both spectra extend to low pulse heights, consistent with Xe-133. The broad peak in the 30-keV spectrum illustrates the difficulties that irradiated xenon may present.

CONCLUSIONS

The production of the fission-produced radioxenons has been successful, and a reliable supply of the short-lived Xe-135 has been demonstrated. Optimization of the process would be the next logical step, and it is foreseen that using other chemical forms of uranium along with more-intense neutron fluencies would significantly enhance this process. The production of short-lived krypton isotopes adds additional energy-calibration points that can be used when calibrating a beta-gamma coincidence detector.

The neutron activation has also been demonstrated and that it is possible, using existing equipment, to produce at least one of the four light radioxenons and Xe-133. The technique has also indicated that additional radioxenon

isotopes may show up in fielded ARSA and SAUNA units in the form of neutron-light radioxenon isotopes. It is important to include these neutron-light radioxenon isotopes as possible contaminants when extra peaks are detected in the beta-gated gamma spectrum. These isotopes have not been seen during the Freiburg tests of Phase II, nor have they been seen during the currently running Phase III tests. However, it is important for IMS analysts and others who will be responsible for the analysis of the radioxenon spectrum from all of the deployed systems to know of the potential signatures that these isotopes may have in some xenon collection units and not confuse the signatures with other fission-produced radioxenon isotopes.

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